Data Testing at INL Using Irradiation Experiments

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Introduction

- The advanced nuclear systems and associated fuel cycles will need good quality cross section data to provide a reliable assessment of their performance.
- Closed fuel cycles with the objective of waste minimization imply, from a physics point of view:
  - A high content of minor actinides in the reactor core and in the fuel cycle
  - A high Fissile/Fertile isotope content in the core fuel
  - A variable, and potentially degraded, Pu isotopic vector in all the fuel cycle
  - Lower fuel density to achieve lower conversion ratios
- Basic data are available for TRU (transuranics) isotopes (up to Cf) but a validation is needed in order to quantify their reliability. The high amount of minor actinides (MA) foreseen in advanced fuel cycle systems requires a specific validation work especially for capture and fission cross sections of such isotopes.

Introduction

- **The validation is traditionally done through the use of differential and integral experiments, and uncertainty assessment.**
- **The information that can be gathered on minor actinides (MA) from experiments comes mostly from small sample irradiation, reactivity oscillation, and fission and capture rates measurements. Separate isotope sample and fuel pin irradiation in power reactors provides a unique source of very useful measurements.**
- **The results of the analysis of such experimental data provide indications to nuclear data evaluators for improving the quality of basic files, and to assess their impact on advanced fuel cycles. These kinds of experiments belong to the category of elemental (separate) effects used in validation methodology.**

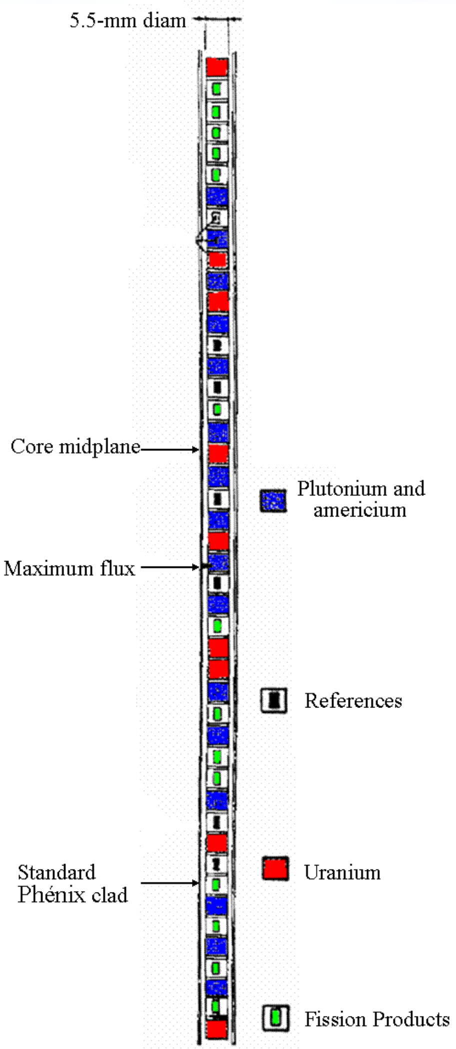
Introduction

- **The experimental data of the PROFIL and TRAPU irradiation experiments , performed at the CEA fast reactor PHENIX, provide very clean and useful information on both cross section data and transmutation rates of actinides.**
- **These data are essential for the validation of the methods and data to be used in advanced fuel cycles where transmuter systems will be used to reduce the existing inventory of nuclear waste. In this presentation these irradiation experiments are used for validating ENDF/B-VII data.**
- **Moreover, in the validation process the use of sensitivity analysis allows to better gather information and indicate specific needs.**

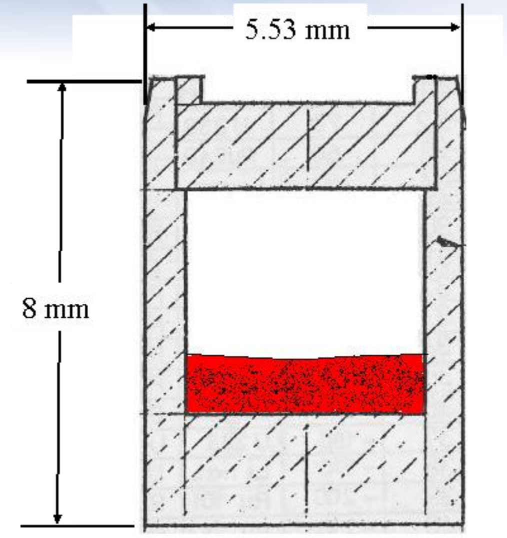
PROFIL-1 Experiment

- **During the PROFIL-1 experiment performed in 1974 a pin containing 46 samples of pure isotopes, including fission products, major and minor actinides (Uranium, Plutonium, and Americium isotopes) was irradiated in the PHENIX fast reactor for the first three cycles, corresponding to a total of 189.2 full-power days.**
- **The experimental pin was located in the central subassembly of the core, and in the third row of pins inside the subassembly. This location is far away from neutronic perturbations allowing clear irradiation conditions.**
- **Following the reactor irradiation, mass spectroscopy was then used, with simple or double isotopic dilution and well-characterized tracers to measure concentrations. The experimental uncertainty obtained with this method was relatively small (maximum of 3%).**

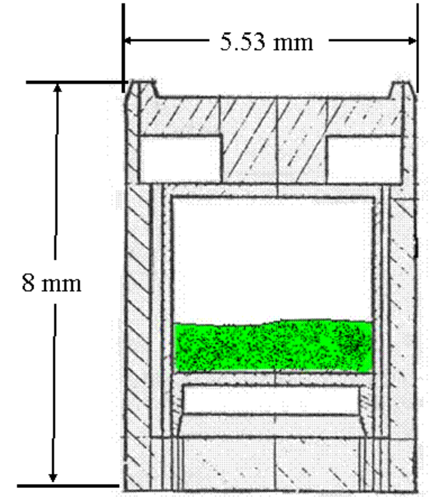
Isotope Sample Irradiation Pin Description



Axial distribution of isotope samples
in irradiation pin



Single-wall capsule for Actinide samples

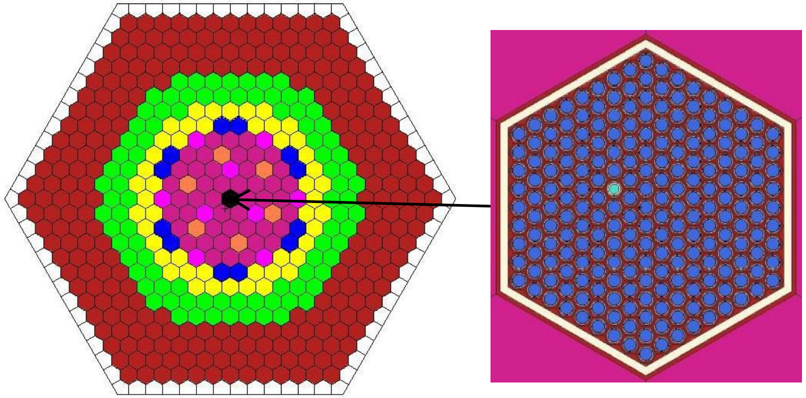


Double-wall capsule for fission product samples

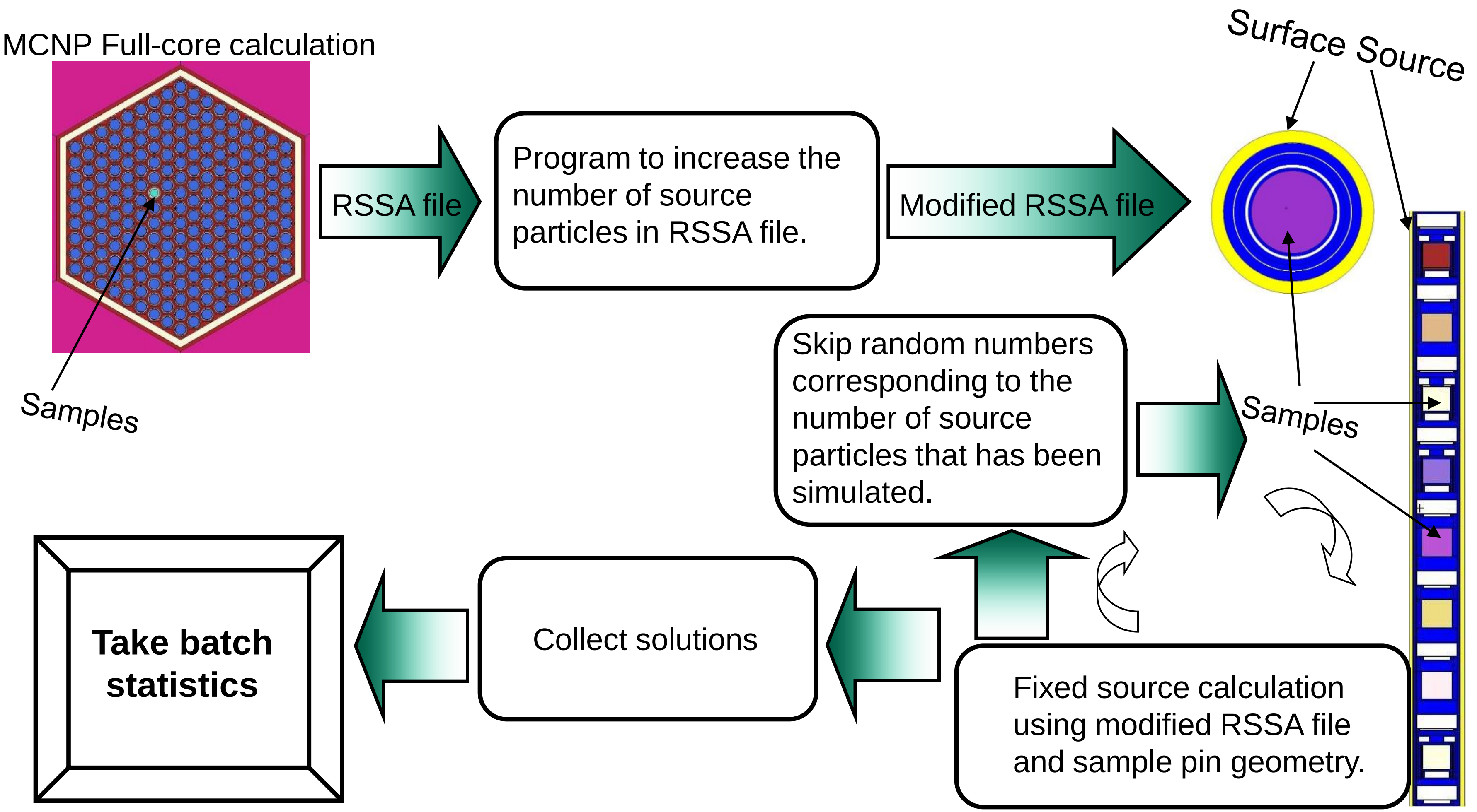
PROFIL-1 Analysis

- MCNP models for PROFIL-1 experiments have been developed.
- One group cross sections for PROFIL-1 samples were calculated by means of solutions obtained by taking the batch statistics of several runs with recorded surface sources.
- For deterministic calculations a full-core VARIANT model and ECCO subassembly model for PROFIL-1 have been developed. The full-core VARIANT calculation has shown very close k_{eff} to that calculated by MCNP.

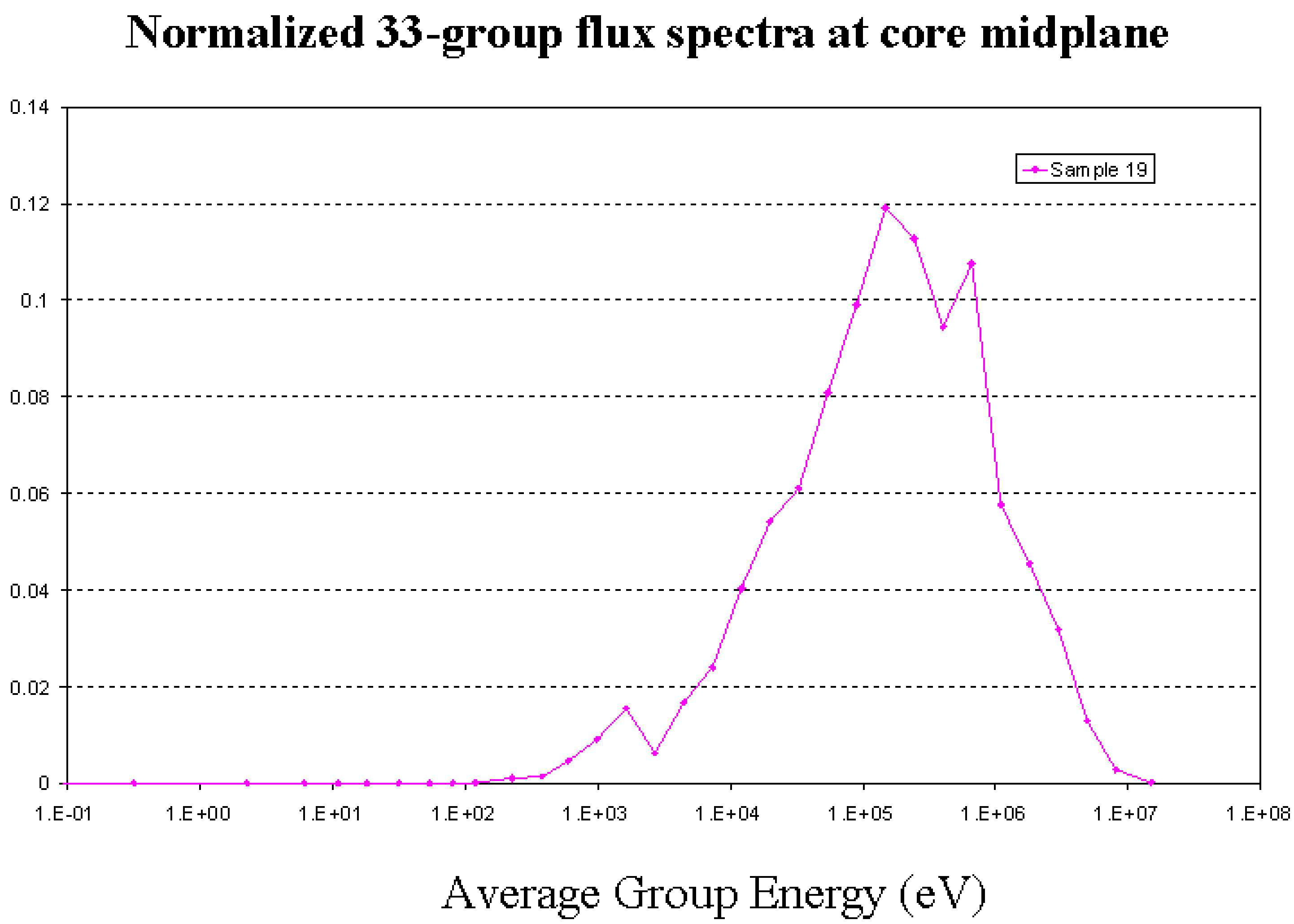
PROFIL-1



Procedure to calculate one-group cross sections over each sample with MCNP



Neutronics Calculation Results



Calculational Scheme

- **One group cross sections were generated with MCNP and with ECCO/VARIANT using ENDF/B-VII files.**
- **In order to correctly normalize the results to the actual value of the flux (and hence eliminate the uncertainty in the total burnup), the production of Nd in the ^{235}U samples has been calculated and compared with the correspondent experimental values. Correcting factors have been obtained and applied to the values of the fluxes used in the time-dependent calculations .**
- **Time dependent calculations were subsequently performed with the NUTS code in order to obtain isotope concentrations at the end of irradiation.**

Calculational Scheme

- **In the past it was used:**

$$\sigma_{(c),A} \cdot \tau \cdot f(\tau) \cong \frac{\Delta N_{A+1}}{N_A} = \frac{N_{A+1}(\tau)}{N_A(\tau)} - \frac{N_{A+1}(0)}{N_A(0)}$$

$$f(\tau) = \left[\frac{N_{A+1}(\tau)}{N_A(\tau)} - \frac{N_{A+1}(0)}{N_A(0)} \right] \times \frac{1}{\sigma_{CA} \cdot \tau_{\mathrm{calc.}}}$$

- **In a more accurate approach we correct the experimental density variation by a calculated quantity that takes out the variation due to all the phenomena other than the reaction rate that we are considering**:

$$\sigma_{(c),A} \cdot \tau \cdot \cong \frac{{}^{corr}\Delta N_{A+1}}{N_A} = \frac{{}^{\mathbf{exp}}\Delta N_{A+1}(\tau) - \left({}^{calc}\Delta N_{A+1} - N_A^{(0)} e^{-\sigma\tau}\right)}{N_A}$$

PROFIL 1 C/E

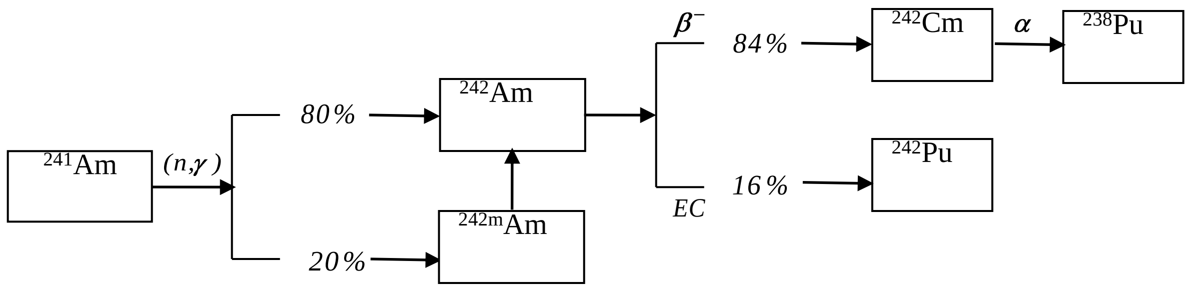
C/E's: Pu242 sample

Samples	Am243	Cm244	Pu242 σ_c	Am243 σ_c
15	1.058	0.826	1.058	0.768
32	1.044	0.933	1.045	0.890
40	1.078	0.925	1.079	0.852

C/E's: Am241 samples

Samples	Pu238	Pu242	Am242	a) Am241 σ_c	b) Am241 σ_c	c) Am241 σ_c
11	0.949	0.965	0.972	0.969	0.965	0.944
44	0.949	0.988	1.001	1.001	0.988	0.945

a) Calculated from Am242 build up. b) Calculated from Pu242 build up. c) Calculated from Pu238 build up



PROFIL-1 one-group cross sections C/E using stochastic and deterministic methods

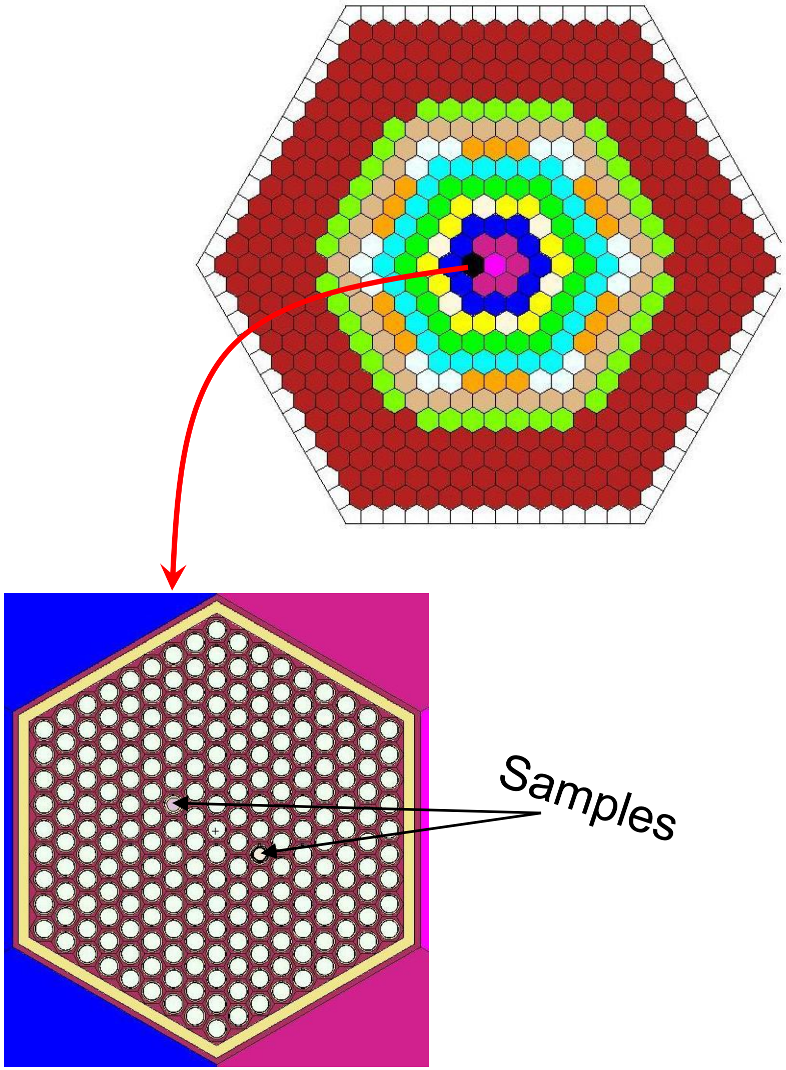
σ	C/E		
	MCNP	Deter.	Exp. Unc.
σ_{capt} U-235	0.948	0.958	1.7 %
σ_{capt} U-238	0.972	0.967	2.3 %
σ_{capt} Pu-238	1.299	1.348	4.0 %
σ_{capt} Pu-239	0.906	0.922	3.0 %
$\sigma_{n,2n}$ Pu-239	0.745	0.663	15.0 %
σ_{capt} Pu-240	0.964	0.976	2.2 %
$\sigma_{n,2n}$ Pu-240	0.779	0.715	15.0 %
σ_{capt} Pu-241	0.950	0.956	4.1 %
σ_{capt} Pu-242	1.061	1.062	3.5 %
σ_{capt} Am-241	0.968	0.975	1.7 %
σ_{capt} Am-243	0.834	0.845	5.0 %
σ_{capt} Mo-95	1.032	1.025	3.8 %
σ_{capt} Mo-97	0.968	0.993	4.4 %
σ_{capt} Ru-101	1.101	1.124	3.6 %
σ_{capt} Pd-105	0.852	0.834	4.0 %
σ_{capt} Cs-133	0.878	0.905	4.7 %
σ_{capt} Nd-145	0.955	1.033	3.8 %
σ_{capt} Sm-149	0.915	0.924	3.1 %

PROFIL-2 Experiment

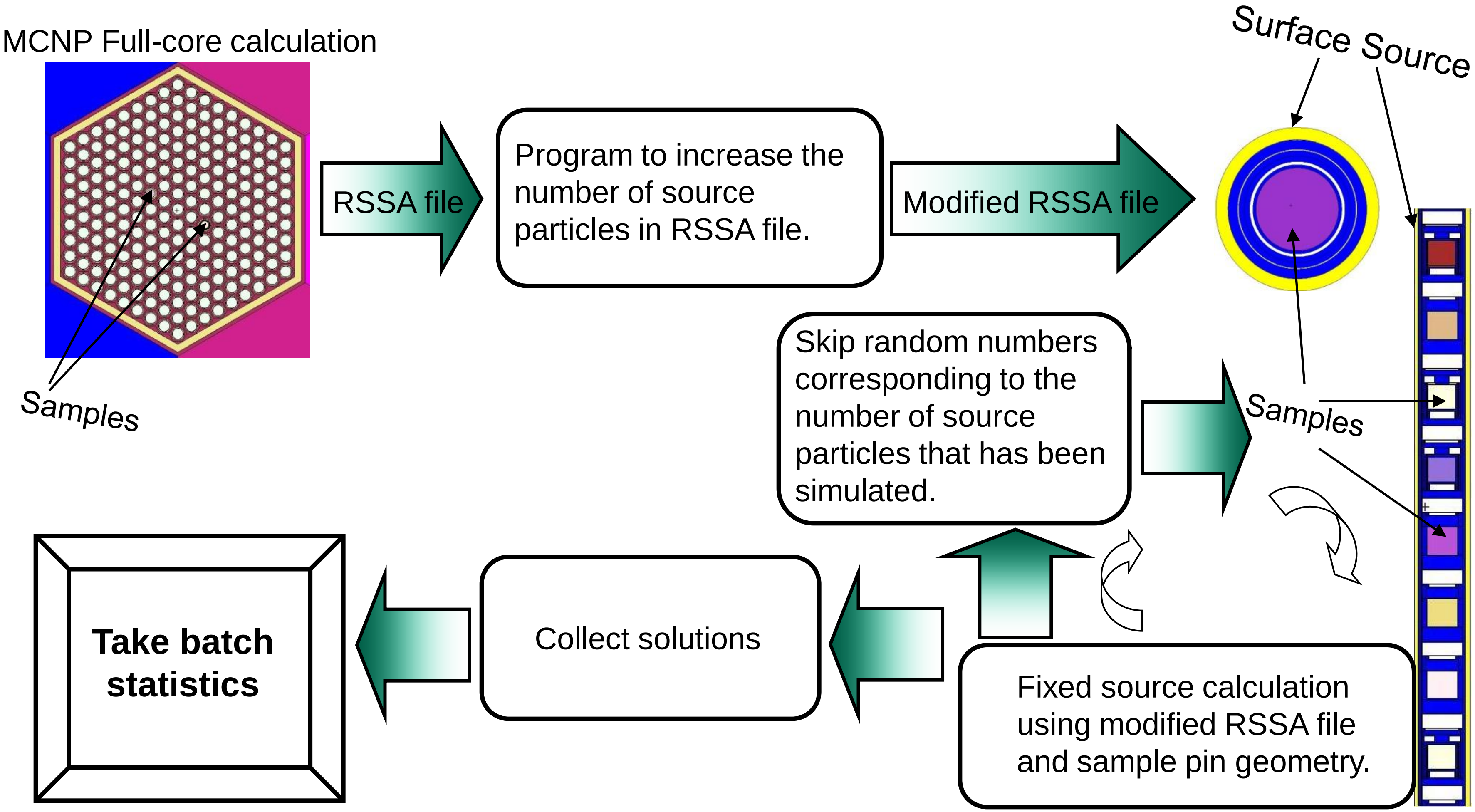
- The second part of the PROFIL irradiation campaign took place in 1979. During the experiment two standard pins, each containing 42 separated capsules of fission products, major and minor actinides (Uranium, Plutonium, Americium and Neptunium isotopes), were irradiated for four cycles (from 17th to 20th) in the fast neutron spectrum reactor PHENIX in France.
- The experimental pins were located in the second row of the core and in the two experimental pins in the third row of the subassembly. Chemical and mass spectrometry analyses have been subsequently performed to determine the post-irradiation isotopic concentrations.

PROFIL-2 Analysis

- Total of 84 samples in two sample pins.
- The same procedure as we have done for PROFIL-1 was performed to generate one-group cross sections.
- Since there are two sample pins, two separate surface source files were generated for fixed source calculations for each pin.



Procedure of One-group Cross Section Generation for PROFIL-2 Experiment



C/E Consistency Between PROFIL-1 and -2

σ	C/E		
	PROFIL-1	PROFIL-2	Exp. Unc.
σ_{capt} U-235	0.948	0.967	1.7 %
σ_{capt} U-238	0.972	0.985	2.3 %
σ_{capt} Pu-238	1.299	1.341	4.0 %
σ_{capt} Pu-239	0.906	0.922	3.0 %
$\sigma_{n,2n}$ Pu-239	0.745	0.574	15.0 %
σ_{capt} Pu-240	0.964	0.973	2.2 %
σ_{capt} Pu-242	1.061	1.054	3.5 %
σ_{capt} Am-241	0.968	1.018	1.7 %

ENDFB-VII.0 and VII.1 C/E for PROFIL-1

σ	C/E		
	VII.0	VII.1	Exp. Unc.
σ_{capt} U-235	0.948	0.948	1.7 %
σ_{capt} U-238	0.972	0.972	2.3 %
σ_{capt} Pu-238	1.299	1.135	4.0 %
σ_{capt} Pu-239	0.906	0.906	3.0 %
$\sigma_{n,2n}$ Pu-239	0.745	0.745	15.0 %
σ_{capt} Pu-240	0.964	0.945	2.2 %
$\sigma_{n,2n}$ Pu-240	0.779	1.084	15.0 %
σ_{capt} Pu-241	0.950	0.947	4.1 %
σ_{capt} Pu-242	1.061	1.120	3.5 %
σ_{capt} Am-241	0.968	0.984	1.7 %
σ_{capt} Am-243	0.834	0.834	5.0 %
σ_{capt} Mo-95	1.032	1.063	3.8 %
σ_{capt} Mo-97	0.968	0.993	4.4 %
σ_{capt} Ru-101	1.101	1.095	3.6 %
σ_{capt} Pd-105	0.852	0.845	4.0 %
σ_{capt} Cs-133	0.878	0.827	4.7 %
σ_{capt} Nd-145	0.955	0.936	3.8 %
σ_{capt} Sm-149	0.915	0.908	3.1 %

Color Code

- **Better**
- **Worse**
- **Equivalent**
- **Need Improvement**

ENDFB-VII.0 and VII.1 C/E for PROFIL-2

Data Type	VII.0	VII.1	Exp. Unc.
σ_{capt} U-235	0.967	0.967	1.7 %
σ_{capt} U-238	0.985	0.985	2.3 %
σ_{capt} Np-237	0.944	0.941	3.6 %
σ_{capt} Pu-238	1.341	1.181	4.0 %
σ_{capt} Pu-239	0.922	0.922	3.0 %
$\sigma_{(n,2n)}$ Pu-239	0.574	0.574	15.0%
σ_{capt} Pu-240	0.973	0.961	2.2 %
σ_{capt} Pu-242	1.054	1.114	4.3 %
σ_{capt} Am-241	1.018	1.029	1.7 %
σ_{capt} Cm-244	1.101	0.956	2.0 %
σ_{capt} Pd-106	0.939	0.939	2.0 %
σ_{capt} Nd-143	0.937	0.937	2.0 %
σ_{capt} Nd-144	0.935	0.928	2.0 %
σ_{capt} Sm-147	0.894	0.894	2.0 %
σ_{capt} Sm-151	1.094	1.085	2.0 %
σ_{capt} Eu-153	0.924	0.954	2.0 %

Color Code

Better

Worse

Equivalent

Need Improvement

TRAPU Experiment

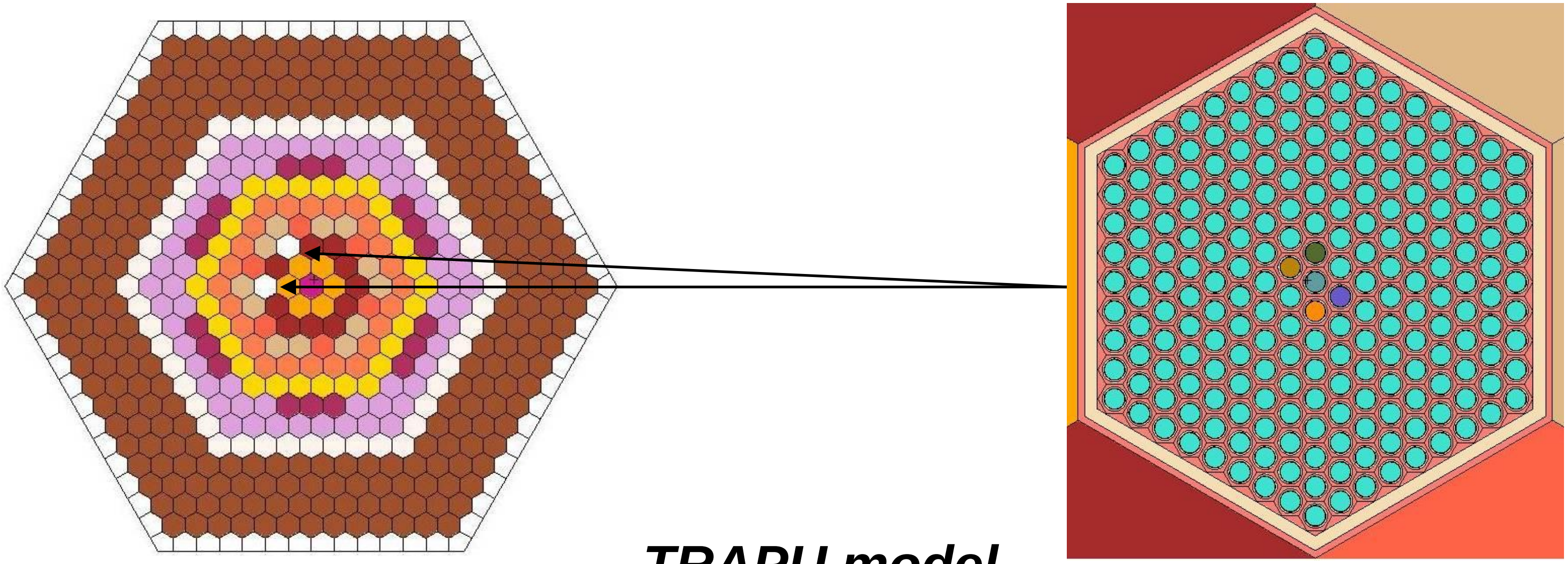
- **The TRAPU experiment consisted of a six-cycle irradiation (10th to 15th) of mixed-oxide pins that contained plutonium of different isotopic compositions but heavily charged in the higher isotopes (Pu240, Pu241 and Pu242) compared to typical PHENIX fuel. Standard pins were placed in regular PHENIX subassemblies located in the third row of the reactor. Three types of plutonium containing pins were used.**
- **After irradiation, small samples (20 mm high) were cut from the experimental pins (both fuel and clad) and put into a solution in order to determine the fuel composition by nuclide. Neodymium-148 was used as burn up indicator since it is a stable fission product with a small capture cross section, and it enables determination of the number of fission reactions that have taken place in the sample. Mass spectrometry was then used, with simple or double isotopic dilution and well-characterized tracers.**

Plutonium Isotopic Compositions of the Three TRAPU Fuel Pins

Pin	Isotope Composition [%]				
	Pu-238	Pu-239	Pu-240	Pu-241	Pu-242
TRAPU-I	0.1	73.3	21.9	4.0	0.7
TRAPU-II	0.8	71.4	18.5	7.4	1.9
TRAPU-III	0.2	34.0	49.4	10.0	6.4

TRAPU Modeling

- ERANOS input for TRAPU developed by CEA has been converted to MCNP model.
- TRAPU has a larger sample volume than PROFIL-1&2. (make MC calculation easier).



TRAPU model

Experimental Concentrations for the TRAPU Program (U238=100).

	TRAPU-I		TRAPU-II		TRAPU-III	
Isotope	Initial	Final	Initial	Final	Initial	Final
U-234	0.0060 ± 3.3 %	0.0062 ± 2.5 %	0.0106 ± 4.7 %	0.01278 ± 1.3 %	0.0088 ± 5.6 %	0.00995 ± 1.0%
U-235	0.7263 ± 0.3 %	0.4830 ± 0.3 %	0.7614 ± 0.3 %	0.4969 ± 0.2 %	0.7447 ± 0.3 %	0.4869 ± 0.2%
U-236	0.0015 ± 13.3 %	0.0664 ± 0.5 %	0.0025 ± 8.0	0.0688 ±0.4 %	0.0099 ± 5.0%	0.07801 ± 0.3%
Np-237	0.0001 ± (*)	0.0365 ± 6.8 %	0.0011 ± (*)	0.0390 ±3.3 %	0.0042 ± (*)	0.0473 ± 3.2%
Pu-238	0.0296 ± 1.3%	0.0455 ± 0.9 %	0.1804 ±0.5 %	0.2020 ±0.4 %	0.0852 ± 0.6%	0.2420 ± 0.4%
Pu-239	17.939 ± 0.5%	16.366 ± 0.4 %	16.780 ± 0.5 %	15.852 ±0.3 %	13.068 ± 0.5%	13.338 ± 0.3%
Pu-240	5.367 ± 0.5%	6.308 ± 0.4 %	4.359 ± 0.5 %	5.433 ± 0.3 %	19.006 ± 0.5%	18.197 ± 0.3%
Pu-241	0.9768 ± 0.5%	0.9449 ± 0.4 %	1.744 ± 0.5 %	1.304 ± 0.3 %	3.858 ± 0.5%	3.406 ± 0.3%
Pu-242	0.1744 ± 0.6%	0.2353 ± 0.5 %	0.4472 ± 0.5 %	0.5473 ±0.4 %	2.455 ± 0.5%	2.598 ± 0.3%
Am-241	0.0657 ± 2.0%	0.1410 ± 3.0 %	0.3432 ± 2.0 %	0.3915 ± 3.6 %	1.029 ± 2.1%	1.084 ± 2.1%
Am-242	-	0.0044 ± 3.6 %	-	0.01437 ± 4.0 %	-	0.0419 ± 12.5%
Am-243	-	0.0160 ±3.6 %	-	0.03945 ± 4.0 %	-	0.1888 ± 2.5%
Cm-242	-	0.00765 ± 2.4 %	-	0.02506 ± 2.6 %	-	0.07052 ± 2.1%
Cm-243	-	-	-	0.002475 ± 2.7 %	-	0.006985 ± 2.6%
Cm-244	-	0.00243 ± 2.4 %	-	0.005366 ± 2.2 %	-	0.02684 ± 1.7%
Nd-148	-	0.1464 ± 0.3 %	-	0.1468 ± 0.2 %	-	0.1720 ± 0.2%

(*) This value has not been measured, but deduced from the Am-241 decay

C/E Values of TRAPU-1

Isotope	TRAPU 1		
	VII.0	VII.1	Exp. Unc.
U-234	1.006	1.004	± 3.9 %
U-235	1.001	1.002	± 0.4%
U-236	0.972	0.971	± 0.8 %
Np-237	0.970	0.879	± 6.8 %
Pu-238	1.004	0.992	± 1.5 %
Pu-239	1.031	1.034	± 0.6 %
Pu-240	1.003	1.007	± 0.6 %
Pu-241	1.011	1.004	± 0.6 %
Pu-242	1.036	1.028	± 0.8 %
Am-241	0.979	0.975	± 3.2 %
Am242M	1.009	1.016	± 3.8 %
Am-243	0.978	1.025	± 2.6 %
Cm-242	1.035	0.984	± 3.9 %
Cm-243	-	-	-
Cm-244	0.843	0.882	± 2.1 %

Color Code

Better

Worse

Equivalent

Need Improvement

C/E Values of TRAPU-2

Isotope	TRAPU 2		
	VII.0	VII.1	Exp. Unc.
U-234	1.023	1.026	± 3.8 %
U-235	1.020	1.021	± 0.4 %
U-236	0.995	0.992	± 1.0 %
Np-237	0.963	0.988	± 3.3 %
Pu-238	0.990	0.998	± 1.0 %
Pu-239	1.012	1.014	± 0.5 %
Pu-240	0.984	0.985	± 0.6 %
Pu-241	0.992	0.988	± 0.6 %
Pu-242	1.010	1.003	± 0.6 %
Am-241	0.986	0.983	± 3.9 %
Am242M	1.039	1.049	± 4.3 %
Am-243	0.959	1.010	± 3.1 %
Cm-242	1.017	0.964	± 3.1 %
Cm-243	0.483	1.104	± 3.1 %
Cm-244	0.946	0.996	± 2.3 %

Color Code

- Better
- Worse
- Equivalent
- Need Improvement

C/E Values of TRAPU-3

Isotope	TRAPU 3		
	VII.0	VII.1	Exp. Unc.
U-234	1.065	1.067	± 4.6 %
U-235	1.019	1.019	± 0.4 %
U-236	0.992	0.991	± 0.9 %
Np-237	0.908	0.915	± 3.2 %
Pu-238	1.013	1.001	± 1.6 %
Pu-239	1.018	1.020	± 0.4 %
Pu-240	0.998	1.002	± 0.6 %
Pu-241	1.004	0.999	± 0.6 %
Pu-242	1.009	1.003	± 0.6 %
Am-241	0.991	0.987	± 2.6 %
Am242M	1.021	1.031	± 3.1%
Am-243	1.000	1.050	± 2.5 %
Cm-242	1.011	0.959	± 2.7 %
Cm-243	0.490	1.106	± 3.2 %
Cm-244	0.961	1.009	± 1.8 %

Color Code

Better

Worse

Equivalent

Need Improvement

Sensitivity (%) to isotope build up: TRAPU-2

Basic Data	Isotope build-up						
	^{234}U	^{235}U	^{236}U	^{237}Np	^{238}Pu	^{239}Pu	^{240}Pu
^{234}U σ_{cap}	-10	0.2					
^{234}U σ_{fis}	-6						
^{235}U σ_{cap}		-10.8	90	9.7	0.2		
^{235}U σ_{fis}	-0.2	-36.2	-16.2	-1.2			
^{236}U σ_{cap}			-5.7	10.5	0.2		
^{238}U σ_{cap}				-2.3		26.7	3.9
^{238}U σ_{fis}				-0.4		-0.1	
^{238}U $\sigma_{(n,2n)}$	0.1			82.5	1.8		
^{237}Np σ_{cap}	0.2			-14.0	1.9		
^{237}Np σ_{fis}				-3.1			
^{238}Pu σ_{cap}	-1.1				-8.0		
^{238}Pu σ_{fis}	-2.1				-16.9		
^{239}Pu σ_{cap}			0.2			-8.1	26.2
^{239}Pu σfis						-29.4	-4.3
^{240}Pu σ_{cap}			-0.1	0.1	0.4		-9.3
^{240}Pu σ_{fis}			-0.1				-6.7
^{241}Pu σ_{cap}					-0.1		
^{241}Pu σ_{fis}				-0.1	-0.6		
^{241}Am σ_{cap}	2.7			-0.7	26.1		

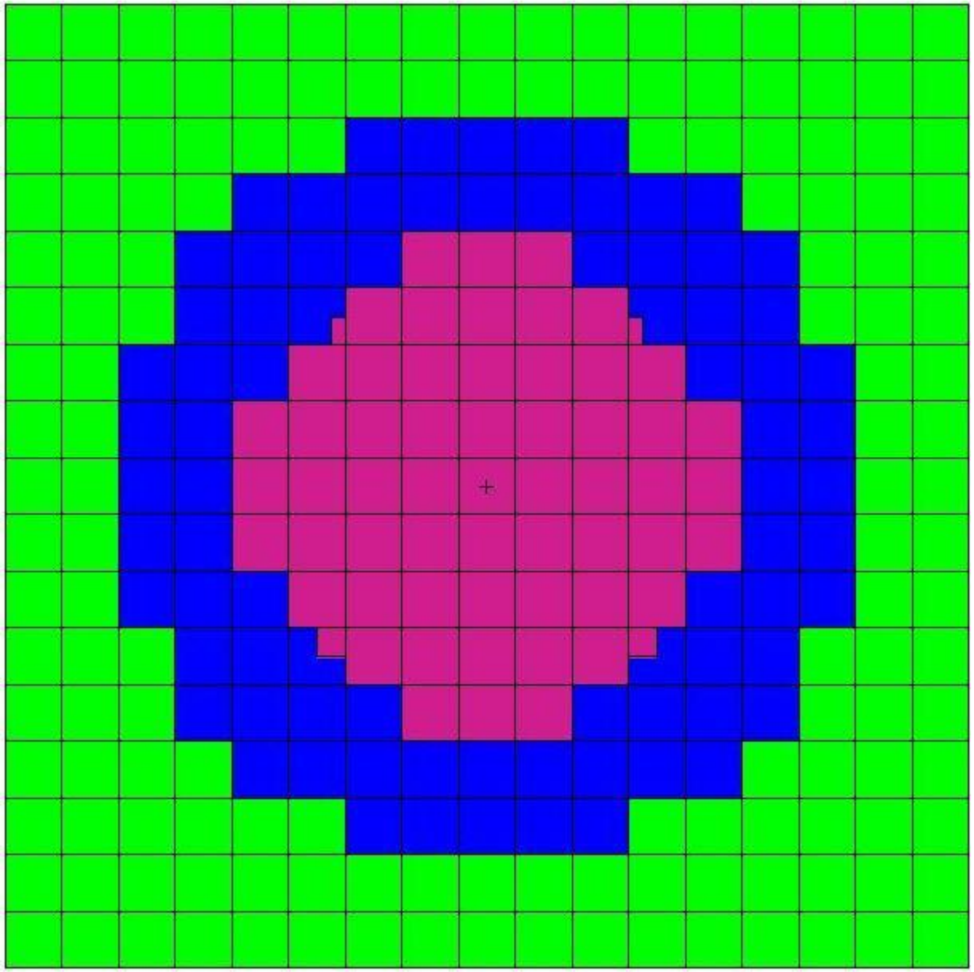
Sensitivity (%) to isotope build up: TRAPU-2

Basic Data	Isotope build-up							
	^{241}Pu	^{242}Pu	^{241}Am	^{242}Am	^{243}Am	^{242}Cm	^{243}Cm	^{244}Cm
^{238}U σ_{cap}	0.6							
^{239}Pu σ_{cap}	5.7	0.4	0.6	0.1	0.2	0.2		
^{239}Pu σfis	-0.7		-0.2					
^{240}Pu σ_{cap}	30.5	3.5	4.4	1.5	1.7	2.0	1.2	0.8
^{240}Pu σ_{fis}	-1.2	-0.1	-0.1					
^{241}Pu σ_{cap}	-8.8	24.2	-1.4	-0.5	15.5	-0.7	-0.4	10.7
^{241}Pu σ_{fis}	-40.7	-5.4	-6.7	-2.4	-2.5	-3.2	-1.8	-1.2
^{242}Pu σ_{cap}		-7.7			93.8		-0.1	95.4
^{242}Pu σ_{fis}		-4.6			-2.4			-1.8
^{241}Am σ_{cap}		2.8	-30.5	82.1	3.3	77.7	85.4	2.8
^{241}Am σ_{fis}			-4.7	-2.8	-0.1	-3.4	-2.2	
^{242}Am σ_{fis}				-27.1	-0.3			-0.2
^{243}Am σ_{cap}					-15.2		0.2	89.7
^{243}Am σ_{fis}					-1.8			-1.4
^{242}Cm σ_{cap}						-3.1	97.7	0.5
^{242}Cm σ_{fis}						-3.7	-2.6	
^{243}Cm σ_{fis}							-20.6	

What about fission cross sections

- The PROFIL and TRAPU experiments can provide information also on fission cross sections; however, the analysis can be tricky. For PROFIL one has to have a good knowledge of the fission yields, and for TRAPU one has to proceed to data adjustment based on sensitivity coefficients.
- A more profitable way to gather information on fission cross sections via elemental experiments is through the analysis of fission spectral indices. In this case, fission reaction rates of actinides are measured against a standard, in particular U-235 fissions. If the measurements are done at the center of the reactor in a very well characterized spectrum, indirect (spectral) effects are normally of the second order and the result can be directly related to the actinide fission cross section.
- This is the case of the COSMO experimental campaign performed at the French zero power fast spectrum facility MASURCA, where different actinide fission spectral indices (with respect to U-235 fissions) were measured.

COSMO Fission Spectral Indices



COSMO C/E

σ	C/E		
	VII.0	VII.1	Exp. Unc.
σ_{fis} U-238	0.984	0.981	1.5 %
σ_{fis} Np-237	1.005	1.004	1.5 %
σ_{fis} Pu-238	1.072	1.040	2.5 %
σ_{fis} Pu-239	0.991	0.989	1.3%
σ_{fis} Pu-240	1.051	1.028	2.3 %
σ_{fis} Pu-241	1.004	1.001	2.0 %
σ_{fis} Pu-242	1.018	1.041	2.3 %
σ_{fis} Am-241	1.089	1.081	2.3 %
σ_{fis} Am-243	1.010	1.009	2.3 %

Color Code

Better

Worse

Equivalent

Need Improvement

Conclusions

- **Elemental experiments like irradiation experiments are extremely useful for validating data related to minor actinides and fission products.**
- **In the PROFIL experiments improvements can be observed for the ENDF/B-VII.1 data for captures of Pu-238, Am-241, Cm244, Mo-97, Sm-151, Eu-153, and (n,2n) of Pu-240. On the other hand, captures Pu-240, Pu-242, Mo-95, Cs-133, and Nd-145 worsen. For major actinides the captures of U-235 and especially Pu-239 remain quite on the underestimated side. Regarding the fission products, Pd-105 and 106, Nd-143 and 144, Sm-147 and 149 are significantly underestimated, while Ru-101 and Sm-151 are overestimated.**
- **From the TRAPU analysis, the major outstanding improvement is related to the build-up of Cm-243, that has to be attributed to a significant amelioration of the Cm-242 capture.**
- **Regarding the fission cross sections, the analysis of the COSMO fission spectral indices has indicated that ENDF/B-VII.1 provides improvements for Pu238 and Pu240 fission cross sections, while Pu-242 worsens. AM241 fission does not change, but it is quite overestimated.**